

Relativistic theory of the double photoionization of helium-like atoms

Vladimir A. Yerokhin^{1,2,3} and Andrey Surzhykov^{1,2}

¹*Institute of Physics, University of Heidelberg, Philosophenweg 12, D-69120 Heidelberg, Germany*

²*GSI Helmholtzzentrum für Schwerionenforschung GmbH, Planckstraße 1, D-64291 Darmstadt, Germany*

³*Center for Advanced Studies, St. Petersburg State Polytechnical University, Polytekhnicheskaya 29, St. Petersburg 195251, Russia*

A fully relativistic calculation of the double photoionization of helium-like atoms is presented. The approach is based on the partial-wave representation of the Dirac continuum states and accounts for the retardation in the electron-electron interaction as well as the higher-order multipoles of the absorbed photon. The electron-electron interaction is taken into account to the leading order of perturbation theory. The relativistic effects are shown to become prominent already for the medium- Z ions, changing the shape and the asymptotic behaviour of the photon energy dependence of the ratio of the double-to-single photoionization cross section.

PACS numbers: 32.80.Fb, 31.15.aj, 33.60.+q, 34.10.+x

I. INTRODUCTION

The ejection of two electrons caused by absorption of a single photon is one of the fundamental few-body processes in atomic physics. The process is called double photoionization or, less frequently, photo-double-ionization. The characteristic feature of this process is that it proceeds exclusively through the electron-electron interaction. Because of this, double photoionization has long been used as a testing ground for understanding of the electron correlation phenomena.

The traditional system for studying double photoionization is the helium atom, for which numerous experimental and theoretical investigations have been performed during the last four decades (for a recent review, see Ref. [1]). Most widely studied is the ratio of the double-to-single photoionization cross sections $R = \sigma^{++}/\sigma^+$ as a function of the energy of the incoming photon ω . Early calculations of this ratio were mainly concentrated either at the near-threshold region of the photon energies ($\omega \gtrsim \omega_{\text{cr}} \approx 79$ eV for helium, with ω_{cr} being the double ionization energy), where Wannier theory is shown to be applicable [2, 3], or at the asymptotical nonrelativistic region $\omega_{\text{cr}} \ll \omega \ll m$ [4–6] (where m is the electron mass). The intermediate region of the photon energies ($\omega \approx 200$ eV for helium, where $R(\omega)$ reaches its maximum) turned out to be much more difficult for an accurate theoretical treatment. Reliable theoretical predictions in this region were obtained only in the end of 1990th by means of sophisticated many-body techniques (notably, the close-coupling methods) [7–10].

Since the helium atom is an essentially nonrelativistic system, any relativistic effects in its double photoionization are considered to be of little importance at the present level of experimental precision. However, the recent experiments on the double K -shell photoionization of moderately heavy atoms up to silver [11–15] demonstrated significant enhancements (of about factor of five for silver) of the cross section as compared with results of nonrelativistic calculations. A natural candidate for

explaining this enhancement would be relativity, which obviously cannot be disregarded when dealing with the deeply bound electron states in silver. Because of the relative isolation of the K -shell electrons from the outer electrons, the double K -shell ionization of a heavy atom is often compared to the double photoionization of the corresponding helium-like ion. However, an accurate theoretical treatment of double K -shell ionization should include both the relativistic effects on the inner-shell electrons and the electron-correlation effects induced by the outer shells. Such a calculation is rather difficult and has not been performed so far.

In contrast to many-electron atoms, the helium-like ion is a relatively simple system for which an *ab initio* description is feasible. So far, there have not been any direct measurements of the double photoionization of helium-like ions. However, with the advent of new powerful light sources, such as the free-electron laser (FLASH) in Hamburg, the Linear Coherent Light Source (LCLS) at Stanford, and the X-ray Free Electron Laser (XFEL) at Hamburg, the experimental study of various photoabsorption processes is going to be possible for a great variety of ions in different charge states [16]. Measurements of the double photoionization in moderately heavy helium-like ions would allow us to effectively test our understanding of the *relativistic* electron-correlation effects.

An alternative approach to the investigation of the double photoionization of highly charged ions is to study it in the inverse kinematics, through the radiative double electron capture. For bare oxygen, such measurement was recently accomplished in Ref. [17]. The experimental upper bound on this process for bare uranium was reported in Ref. [18].

The goal of the present investigation is to perform an *ab initio* relativistic calculation of the double photoionization of a helium-like atom. The electron-electron correlation (both on the initial and the final states) will be taken into account to the leading order of the perturbation theory. To the given order of perturbation theory, the treatment is *exact*, i.e., includes all multipoles of the

absorbed photon, the retardation in the electron-electron interaction as well as the interaction of the electrons with the nucleus without any expansion in the binding field. The treatment is gauge invariant, both with respect to the gauge used for the absorbed photon and the gauge of the electron-electron interaction. The higher-order electron-correlation effects omitted are estimated by comparing the present numerical results with the experimental data available for helium.

The remaining paper is organized as follows. In Sec. II we present a short summary of the relativistic formulas for the single photoionization, which forms the basis for our treatment of the double photoionization. In Sec. III we describe the QED theory of the double photoionization to the lowest relevant order of the perturbation theory. Sec. IV presents details of the calculation. Numerical results are presented and discussed in Sec. V.

The relativistic units ($\hbar = c = m = 1$) are used throughout this paper.

II. SINGLE PHOTOIONIZATION

In this section we summarize the relativistic formulas for the single photoionization, as they build the basis for the description of the double photoionization.

Differential cross section of the photoionization of a one-electron atom is

$$d\sigma^+ = \frac{4\pi^2\alpha}{\omega} |\tau_{fi}^+|^2 \delta(\varepsilon_i + \omega - \varepsilon) d\mathbf{p}, \quad (1)$$

where α is the fine-structure constant, ω is the energy of the absorbed photon, ε_i is the initial (bound-state) energy, ε and \mathbf{p} are the energy and the momentum of the emitted electron, respectively, $\varepsilon = \sqrt{m^2 + \mathbf{p}^2}$. The amplitude of the process τ_{fi}^+ is given by

$$\tau_{fi}^+ = \langle \mathbf{p}m | R_\lambda | \kappa_i \mu_i \rangle, \quad (2)$$

where $|\kappa_i \mu_i\rangle$ denotes the initial Dirac bound state with the relativistic angular quantum number κ_i and its projection μ_i , and $|\mathbf{p}m\rangle$ is the wave function of the emitted electron. The general relativistic expression for the photon absorption operator R_λ is

$$R_\lambda = \boldsymbol{\alpha} \cdot \hat{\mathbf{u}}_\lambda e^{i\mathbf{k} \cdot \mathbf{r}} + G(\boldsymbol{\alpha} \cdot \hat{\mathbf{k}} - 1) e^{i\mathbf{k} \cdot \mathbf{r}}, \quad (3)$$

where $\boldsymbol{\alpha}$ is a three-component vector of the Dirac matrices, $\hat{\mathbf{u}}_\lambda$ is the polarization vector of the absorbed photon, \mathbf{k} is the photon momentum, $\hat{\mathbf{k}} = \mathbf{k}/|\mathbf{k}|$, and G is the gauge parameter. In our treatment, all electron states are the eigenfunctions of the Dirac Hamiltonian, so the gauge-dependent part of R_λ vanishes identically. However, we keep the general gauge of the absorbed photon in actual calculations in order to check the numerical procedure.

The wave function $|\mathbf{p}m\rangle \equiv |\mathbf{p}m\rangle_-$ in Eq. (2) is the one-electron continuum Dirac state with the asymptotic

momentum \mathbf{p} , helicity $m = \pm 1/2$, and the “−” asymptotic behaviour (i.e., the plane wave modified by the Coulomb logarithmic phase plus the incoming spherical wave) [19]

$$|\mathbf{p}m\rangle_- = \frac{u(\mathbf{p}, m)}{(2\pi)^{3/2}} e^{i[pz - \eta \ln p(r-z)]} + f(\theta, \phi) \frac{e^{-i[pr + \eta \ln 2pr]}}{r} \quad (4)$$

as $|r - z| \rightarrow \infty$, where $\eta = Z\alpha\varepsilon/p$ is the Sommerfeld parameter. The free-electron 4-spinors $u(\mathbf{p}, m)$ [20] are normalized by the condition $\bar{u}u \equiv u^\dagger \gamma_0 u = 1$. The explicit expression of the electron wave function is

$$|\mathbf{p}m\rangle_- = \frac{1}{\sqrt{p\varepsilon}} \sum_{\kappa\mu} i^l e^{-i\Delta_l} C_{l m_l, \frac{1}{2}m}^{j\mu} Y_{l m_l}^*(\hat{\mathbf{p}}) |\varepsilon\kappa\mu\rangle, \quad (5)$$

where Δ_l are the scattering phases [21], $j = |\kappa| - 1/2$, $l = |\kappa + 1/2| - 1/2$, and $|\varepsilon\kappa\mu\rangle$ are Dirac continuum states with given relativistic angular momentum κ and the momentum projection μ , normalized on the energy scale [19].

Taking into account that $d\mathbf{p} = p\varepsilon d\varepsilon d\Omega$, the single differential cross section of the photoionization of a hydrogen-like atom is written as

$$\frac{d\sigma^+}{d\Omega} = \frac{4\pi^2\alpha}{\omega} p\varepsilon |\tau_{fi}^+|^2, \quad (6)$$

where the energy of the emitted electron is fixed by the energy conservation, $\varepsilon = \varepsilon_i + \omega$.

In our analysis of the double photoionization, we will need the cross section of the single photoionization of a *helium-like* atom. In the independent-particle approximation, the wave function of the initial two-electron state is

$$|J_0 M_0\rangle = N \sum_{\mu_a \mu_b} C_{j_a \mu_a, j_b \mu_b}^{J_0 M_0} \times \frac{1}{\sqrt{2}} (|\kappa_a \mu_a\rangle |\kappa_b \mu_b\rangle - |\kappa_b \mu_b\rangle |\kappa_a \mu_a\rangle), \quad (7)$$

where $N = 1/\sqrt{2}$ for the equivalent electrons and $N = 1$ otherwise. Averaging the cross section over the momentum projection of the initial state M_0 , employing the explicit expression for the continuum Dirac state (5), and integrating over the angles over the emitted electron, we obtain the total photoionization cross section of a helium-like atom as

$$\sigma^+ = \frac{4\pi^2\alpha}{\omega} \left[\frac{1}{2j_a + 1} \sum_{\kappa\mu\mu_a} |\langle \varepsilon_1 \kappa\mu | R_\lambda(\omega) | \kappa_a \mu_a \rangle|^2 + \frac{1}{2j_b + 1} \sum_{\kappa\mu\mu_b} |\langle \varepsilon_2 \kappa\mu | R_\lambda(\omega) | \kappa_b \mu_b \rangle|^2 \right], \quad (8)$$

where $\varepsilon_1 = \varepsilon_a + \omega$ and $\varepsilon_2 = \varepsilon_b + \omega$ and we assumed that the energy of the photon is sufficient to ionize any of the two initial-state electrons. If this is not the case, only one of the two terms in the brackets should be retained.

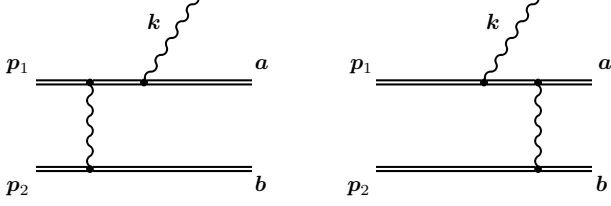


FIG. 1: Feynman diagrams representing the double photoionization of a helium-like atom. a and b denote the bound electron states, p_1 and p_2 are the continuum electron states, k denotes the incoming photon. Double lines denote electrons propagating in the binding nuclear field.

The electron correlation effects on σ^+ omitted in the independent-particle approximation are well studied in the literature. In particular, the leading nonrelativistic term of the $1/Z$ expansion of the high-energy asymptotics of σ^+ was obtained in Ref. [22]. However, since we are presently interested in the ratio of the double-to-single photoionization cross sections σ^{++}/σ^+ , we prefer to treat both these cross sections on the same footing, i.e., to the leading nonvanishing order of perturbation theory with respect to the electron-electron interaction.

III. DOUBLE PHOTOIONIZATION: GENERAL FORMULAS

According to the general rules of quantum field theory [23], the differential cross section of the double photoionization is

$$d\sigma^{++} = \frac{4\pi^2\alpha}{\omega} \left| \tau_{fi}^{++} \right|^2 \delta(\varepsilon_i + \omega - \varepsilon_1 - \varepsilon_2) d\mathbf{p}_1 d\mathbf{p}_2, \quad (9)$$

where $(\varepsilon_1, \mathbf{p}_1)$ and $(\varepsilon_2, \mathbf{p}_2)$ are the energy and the momentum of the two final-state electrons, respectively, ε_i is the energy of the initial bound state, and τ_{fi}^{++} is the amplitude of the process. To the leading order of perturbation theory, the amplitude of the process is represented by the two Feynman diagrams shown in Fig. 1, where the antisymmetrization of the initial and the final two-electron states is assumed.

The initial-state wave function is the same as for the single photoionization [see Eq. (7)] and the initial-state energy is $\varepsilon_i = \varepsilon_a + \varepsilon_b$. The wave function of the final state is

$$\begin{aligned} |p_1 m_1, p_2 m_2\rangle_- &= \frac{1}{\sqrt{2}} \left(|p_1 m_1\rangle_- |p_2 m_2\rangle_- \right. \\ &\quad \left. - |p_2 m_2\rangle_- |p_1 m_1\rangle_- \right). \end{aligned} \quad (10)$$

Note that in the present approach, we use the wave functions that do not include any electron-correlation effects (apart from the antisymmetrization); the electron-electron interaction enters explicitly into the amplitude of the process [see Eq. (16) below]. In the nonrelativistic case, this approach has been successfully applied to

the double photoionization by many authors, notably in Refs. [4, 24]. Although the formal parameter of the perturbative expansion is $1/Z$, the leading-term approximation was shown to adequately describe the double photoionization even for helium. Our present treatment is aimed primarily at the helium-like ions, for which the perturbative expansion converges much faster than for helium.

The triple differential cross section of the double photoionization obtained from Eq. (9) is

$$\begin{aligned} \frac{d^3\sigma^{++}}{d\Omega_1 d\Omega_2 d\varepsilon_1} &= \frac{4\pi^2\alpha}{\omega} p_1 p_2 \varepsilon_1 \varepsilon_2 \\ &\times \sum_{m_1 m_2} \left| \tau_{\lambda}^{++}(\mathbf{p}_1 m_1, \mathbf{p}_2 m_2; \omega, J_0 M_0) \right|^2, \end{aligned} \quad (11)$$

where the energy of the second electron is fixed by the energy conservation, $\varepsilon_2 = \varepsilon_a + \varepsilon_b + \omega - \varepsilon_1$. The energy distribution of the emitted electrons can be conveniently parameterized by the fractional energy sharing parameter x ,

$$\varepsilon_1 = m + x(\omega - \omega_{\text{cr}}), \quad (12)$$

$$\varepsilon_2 = m + (1 - x)(\omega - \omega_{\text{cr}}), \quad (13)$$

where ω_{cr} is the threshold value of the photon energy (the double ionization energy). In our approximation, $\omega_{\text{cr}} = 2m - \varepsilon_a - \varepsilon_b$.

Let us now consider the *single differential cross section* $d\sigma^{++}/d\varepsilon_1$. Substituting the explicit expression for the continuum wave function (5) into Eq. (11), integrating over the angles, and summing over m_1 and m_2 , we arrive at

$$\frac{d\sigma^{++}}{d\varepsilon_1} = \frac{4\pi^2\alpha}{\omega} \sum_{\kappa_1 \kappa_2 \mu_1 \mu_2} \left| \tau_{\lambda}^{++}(\varepsilon_1 \kappa_1 \mu_1, \varepsilon_2 \kappa_2 \mu_2; \omega, J_0 M_0) \right|^2, \quad (14)$$

where the amplitude contains only the spherical-wave Dirac continuum states.

The *total cross section* is obtained as an integral of the single differential cross section over a half of the energy sharing interval

$$\begin{aligned} \sigma^{++} &= \int_m^{m+(\omega-\omega_{\text{cr}})/2} d\varepsilon_1 \frac{d\sigma^{++}}{d\varepsilon_1} \\ &= (\omega - \omega_{\text{cr}}) \int_0^{\frac{1}{2}} dx \frac{d\sigma^{++}}{d\varepsilon_1}. \end{aligned} \quad (15)$$

The other half of the energy interval corresponds to interchanging the first and the second electron, which is already accounted for by the wave function.

The general expression for the transition amplitude of the double photoionization process can be obtained by the two-time Green's function method [23],

$$\begin{aligned} \tau_{\lambda}^{++}(\varepsilon_1 \kappa_1 \mu_1, \varepsilon_2 \kappa_2 \mu_2; \omega, J_0 M_0) = & N \sum_{\mu_a \mu_b} C_{j_a \mu_a j_b \mu_b}^{J_0 M_0} \sum_{PQ} (-1)^{P+Q} \\ & \times \sum_n \left\{ \frac{\langle P \varepsilon_1 P \varepsilon_2 | I(\Delta_{P \varepsilon_2} Q b) | n Q b \rangle \langle n | R_{\lambda} | Q a \rangle}{\varepsilon_{Qa} + \omega - \varepsilon_n (1 - i0)} + \frac{\langle P \varepsilon_1 | R_{\lambda} | n \rangle \langle n P \varepsilon_2 | I(\Delta_{P \varepsilon_2} Q b) | Q a Q b \rangle}{\varepsilon_{P \varepsilon_1} - \omega - \varepsilon_n (1 - i0)} \right\}. \end{aligned} \quad (16)$$

The first term in the brackets corresponds to the electron-electron interaction modifying the final-state electron wave function (the left graph in Fig. 1) and the second one, the initial-state electron wave function (the right graph in Fig. 1). The summation over P and Q corresponds to the permutation of the initial and final electrons, $P \varepsilon_1 P \varepsilon_2 = (\varepsilon_1 \varepsilon_2)$ or $(\varepsilon_2 \varepsilon_1)$, $Q a Q b = (ab)$ or (ba) , and $(-1)^P$ and $(-1)^Q$ are the permutations sign. The summation over n in Eq. (16) runs over the complete Dirac spectrum, $\Delta_{ab} \equiv \varepsilon_a - \varepsilon_b$, and $I(\Delta)$ is the electron-electron interaction operator,

$$I(\omega) = e^2 \alpha_{\mu} \alpha_{\nu} D^{\mu\nu}(\omega, \mathbf{x}_{12}), \quad (17)$$

where $D^{\mu\nu}$ is the photon propagator, $\alpha_{\mu} = (1, \boldsymbol{\alpha})$ are the Dirac matrices. In the Feynman gauge, the electron-electron interaction takes the form

$$I^{\text{Feyn}}(\omega) = \alpha (1 - \boldsymbol{\alpha}_1 \cdot \boldsymbol{\alpha}_2) \frac{e^{i|\omega| x_{12}}}{x_{12}}, \quad (18)$$

where $x_{12} = |\mathbf{x}_1 - \mathbf{x}_2|$. In the Coulomb gauge, the electron-electron interaction acquires an additional term, which can be expressed as

$$\begin{aligned} I^{\text{Coul}}(\omega) = & I^{\text{Feyn}}(\omega) \\ & + \alpha \left[1 - \frac{(\boldsymbol{\alpha}_1 \cdot \boldsymbol{\nabla}_1)(\boldsymbol{\alpha}_2 \cdot \boldsymbol{\nabla}_2)}{\omega^2} \right] \frac{1 - e^{i|\omega| x_{12}}}{x_{12}}. \end{aligned} \quad (19)$$

IV. NUMERICAL CALCULATION

The final formulas for the single and double photoionization cross sections are presented in Appendices A and B. The calculation of the single photoionization cross section is straightforward, in contrast to that of the double photoionization. The major difficulty in the numerical evaluation of the double photoionization is the summation over the complete spectrum of the Dirac equation. In this work we use the approach based on the analytical representation of the Dirac Coulomb Green function, which is represented by an infinite sum over the partial waves. The numerical approach was developed in the previous works [25–27], where it was used for calculating the QED and electron-electron interaction corrections to the radiative recombination of electrons with highly charged ions.

For a given value of the relativistic angular momentum quantum number κ , the radial part of the Dirac Coulomb Green function is represented in terms of the two-component solutions of the radial Dirac equation regular at the origin (ϕ_{κ}^0) and the infinity (ϕ_{κ}^{∞}),

$$\begin{aligned} G_{\kappa}(E, r_1, r_2) = & -\phi_{\kappa}^{\infty}(E, r_1) \phi_{\kappa}^{0^T}(E, r_2) \theta(r_1 - r_2) \\ & - \phi_{\kappa}^0(E, r_1) \phi_{\kappa}^{\infty^T}(E, r_2) \theta(r_2 - r_1), \end{aligned} \quad (20)$$

where E denotes the energy argument of the Green function, r_1 and r_2 are the radial arguments, and θ is the step function. For the point Coulomb potential, the regular and irregular solutions of the radial Dirac equation are expressed analytically in terms of the Whittaker functions of the first and second kind (for explicit formulas see, e.g., Ref. [28]).

When the energy argument E is real and greater than the electron rest mass $E > m$, the Dirac Green function is a complex multi-valued function. So, care should be taken in this case to choose the correct branch (i.e., the sign of the imaginary part) of the Green function. The branch of the Green function is fixed by the sign of the infinitesimal imaginary addition $i0$ in the energy denominators of Eq. (16). Specifically, the energy argument of the Green function in the first term in the brackets of Eq. (16), $\varepsilon_{Qa} + \omega$, is greater than the electron mass. For positive-energy intermediate states, the energy denominator takes the form $\varepsilon_{Qa} + \omega - \varepsilon_n + i0$, which implies that the energy argument of the Green function has a small positive imaginary addition and, therefore, the branch cut of the Green function $[m, \infty)$ should be approached from above.

A serious problem arises in the numerical evaluation of the radial integrals for the left graph in Fig. 1 (with the electron correlation modifying the final-state wave function). In this case, the continuum-state Dirac wave function has to be integrated together with the Dirac Green function with the energy argument $E > m$ and the spherical Bessel function. All three functions are strongly oscillating and slowly decreasing for large radial arguments. It is practically impossible to accurately evaluate such integral by a straightforward numerical integration. In this work, we use the method of the complex-plane rotation of the integration contour, which was previously applied by us to the evaluation of the free-free transition integrals in the bremsstrahlung [29].

We now consider the evaluation of the problematic radial integrals in more details. Let us introduce the radius of the atom \mathcal{R} , which is defined as the smallest distance at which all the bound-state electron wave functions vanish. In the inner region $r < \mathcal{R}$, evaluation of each partial-wave expansion term of the amplitude [Eq. (16)] involves a three-dimensional radial integration. In the outer region $r > \mathcal{R}$, however, all the integrals involving the bound-state wave functions reach their asymptotical values and only an one-dimensional integral of the free-free type needs to be evaluated. The general form of such integral is

$$J_{\mathcal{R}} = \int_{\mathcal{R}}^{\infty} dr r^2 j_L(k_3 r) f_{\kappa_2}^i(E_2, r) \phi_{\kappa_1}^{\infty j}(E_1, r), \quad (21)$$

where j_L is the spherical Bessel function originating from the photon propagator in the electron-electron interaction, f^i is the radial component of the Dirac continuum wave function ($i = 1, 2$), and $\phi^{\infty j}$ is the radial component of the irregular solution of the Dirac equation originating from the Green function ($j = 1, 2$). The energy conservation requires that $E_1 = E_2 + k_3$, which leads to the inequality $p_1 > p_2 + k_3$, where $p_{1,2} = \sqrt{E_{1,2}^2 - m^2}$ are the electron momenta associated with the corresponding energies. We now analytically continue the integrand of Eq. (21) into the complex r plane and take into account the asymptotical behaviour of individual functions for large values of $\text{Im}(r)$,

$$f_{\kappa_2}(E_2, r) \sim \exp[p_2 |\text{Im}(r)|], \quad (22)$$

$$\phi_{\kappa_1}^{\infty}(E_1, r) \sim \exp[p_1 \text{Im}(r)], \quad (23)$$

$$j_L(k_3 r) \sim \exp[k_3 |\text{Im}(r)|], \quad (24)$$

where only the leading exponential behaviour is kept. We observe that, rotating the integration contour in the integral (21) into the lower complex half-plane $r \rightarrow -ir$, we transform the strongly oscillating integrand into an exponentially decreasing one, which falls off as $\exp[-(p_1 - p_2 - k_3)r]$ for large r . After the rotation of the contour, the integral can be easily evaluated numerically up to a desired precision.

After the radial integrals are successfully evaluated, the remaining problem is the summation of the partial-wave expansions. Altogether there are five partial-wave expansions to be dealt with: the expansions of the both final electron wave functions, the Green function, the wave function of the absorbed photon, and the photon propagator of the electron-electron interaction. After the angular momentum selection rules are taken into account, only two partial-wave expansions of the two final-state electrons remain unbound. The corresponding expansion parameters are κ_1 and κ_2 in Eq. (14). The convergence of the resulting double partial-wave expansion is good for the photon energies near the threshold $\omega \sim \omega_{\text{cr}}$ and a non-symmetric energy sharing $x \ll 1/2$ but gradually deteriorates when ω increases and x approaches $1/2$. In the most difficult case considered here, $\omega/\omega_{\text{cr}} \approx 30$ and

$x = 1/2$, up to $(|\kappa_1|, |\kappa_2|) = (15, 15)$ partial waves were included into the calculation. The truncated partial wave expansion was extrapolated into infinity by using the ϵ resummation algorithm (see, e.g., Ref. [30]).

In order to check the numerical procedure, we evaluated the nonrelativistic limit of our calculations. The easiest way to do this is to decrease the value of the fine structure constant (or, equivalently, to increase the value of the speed of light) by a large factor. However, a straightforward implementation of this scheme leads to numerical instabilities, since the relativistic operators involve products of the upper and the lower components of the Dirac wave function and the lower component vanishes in the nonrelativistic limit. In order to avoid this problem, we express the lower component in terms of the upper component as

$$f_{\kappa}(r) = \frac{1}{2m} \left(\frac{d}{dr} + \frac{1+\kappa}{r} \right) g_{\kappa}(r), \quad (25)$$

which is valid to the leading order in $Z\alpha$. After this substitution, our relativistic code yielded a stable numerical limit when the fine structure constant was decreased by orders of magnitude, thus giving us the nonrelativistic limit.

V. RESULTS AND DISCUSSION

We start this section with a discussion of the total cross section of the double photoionization. As explained previously, our approach accounts for the electron correlation to the first order of perturbation theory. Within this approximation we perform a rigorous relativistic treatment, without any further simplifications. This approach is expected to yield accurate results for heavy helium-like ions, whereas for light ions, the precision of the method gradually deteriorates with decrease of the nuclear charge. For helium, the relativistic effects are weak but the electron correlation is strong, so that in this case our approach predictably yields worse results than the modern nonrelativistic methods. However, we will use the helium case for estimating the higher-order electron correlation effects omitted, having in mind that in helium-like ions these effects are suppressed by the inverse power of the nuclear charge.

In Fig. 2, we compare the nonrelativistic limit of our calculations of the ratio of the double-to-single photoionization cross section $R(\omega) \equiv \sigma^{++}/\sigma^{+}$ in helium-like ions with the experimental data available for helium and with the theoretical high-energy limits. In order to facilitate the analysis of the results, we plot the scaled ratio $Z^2 R(\omega)$ as a function of the ratio $\omega/\omega_{\text{cr}}$.

Firstly, we confirm the known statement [31, 32] that the *nonrelativistic limit* of the scaled ratio $\bar{R}(\omega) \equiv Z^2 R(\omega/\omega_{\text{cr}})$ is well described by a universal function that does not depend on the nuclear charge number. We will show later that this universal scaling is strongly violated

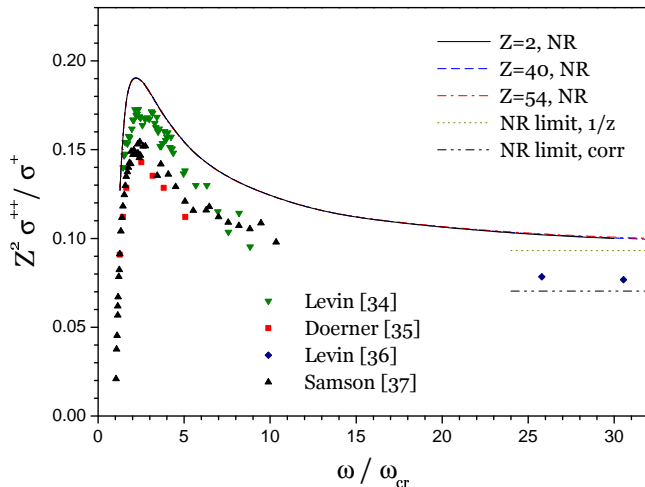


FIG. 2: (Color online) Comparison of the nonrelativistic limit of the present calculations with the experimental results for helium and the nonrelativistic theoretical high-energy limits. The solid (black) line shows our nonrelativistic results for $Z = 2$; the dashed (blue) line, for $Z = 40$; the dashed-dotted (red) line, for $Z = 54$. (The three lines are practically indistinguishable on the picture.) The triangles, squares, and diamonds represent the experimental results for helium obtained in Refs. [34–37], respectively. The dotted line shows the nonrelativistic high-energy limit calculated to the leading order of the perturbation theory [4]. The dash-dot-dotted line shows the nonrelativistic asymptotical limit calculated with the fully correlated wave functions [5, 6].

by the relativistic effects. Fig. 2 shows that the numerical results calculated for different nuclear charges are practically indistinguishable from each other in the nonrelativistic limit.

Secondly, we observe good agreement between our nonrelativistic results and the leading term of the $1/Z$ expansion of the asymptotics [4, 33], $R_{0,1/Z} = 0.0932/Z^2$. The deviation of our numerical results from the experimental data [34–37] is consistent with the deviation of the perturbative asymptotical value $R_{0,1/Z}$ from the fully correlated result $R_{0,corr} = 0.0658/Z^2$ [5, 6]. We, therefore, estimate the higher-order electron-correlation effects omitted in the present treatment to be about $2/Z \times 30\%$ for the ratio $R(\omega)$.

We now turn to our relativistic calculations. Fig. 3 presents our numerical results obtained with a step-by-step inclusion of individual relativistic effects for several helium-like ions. The four different treatments compared are (i) the nonrelativistic calculation (dotted line), (ii) the calculation with the relativistic wave functions but with neglecting the retardation in the electron-electron interaction and all multipoles of the absorbed photon higher than the relativistic $E1$ transition (dash-dotted line), (iii) the calculation with the relativistic wave functions and the full retardation but without the higher multipoles of the absorbed photon (dashed line), and (iv) the

full relativistic treatment (solid line).

We observe that for moderate photon energies, $\omega/\omega_{cr} \lesssim 3$, the dominant relativistic effect is the retardation in the electron-electron interaction, whereas for high photon energies, $\omega/\omega_{cr} \gtrsim 10$, the effect of the higher multipoles of the absorbed photon becomes dominant. Altogether, the relativistic effects are large and change qualitatively the shape of the energy dependence of the ratio $R(\omega)$ already for medium- Z ions. In particular, the maximum of the curve located for helium at $\omega/\omega_{cr} \approx 2.5$ disappears for $Z > 20$ and $R(\omega)$ becomes a monotonically growing function (at least, up to the maximal photon energies accessible in our calculations), which is in contrast to the nonrelativistic case where $R(\omega)$ gradually decreases to approach a constant high-energy limit from above. It is interesting to note that similarly strong relativistic effects were recently reported for the nonelastic electron scattering from the hydrogen-like ions [38]. Significant effects caused by the magnetic dipole and electric quadrupole transitions in the double photoionization of helium-like ions were previously reported in Ref. [39].

In order to check our relativistic calculations, we employed different gauges for the absorbed photon and, separately, for the photon propagator of the electron-electron interaction. We found that the gauge-dependent term in the photon absorption operator [see Eq. (3)] vanishes in the actual calculations. Independently, we demonstrated that our calculations with the electron-electron interaction operator in the Feynman and the Coulomb gauges [see Eqs. (18) and (19)] yield the same results. This was an important cross-check of the numerical procedure since the contributions of the two diagrams in Fig. 1 are separately not gauge invariant. As an additional test, we calculated the contribution of the right graph in Fig. 1 independently by a different numerical technique based on the B -spline representation of the Dirac Coulomb Green function [40].

The results of our relativistic calculations for various helium-like ions are summarized in Fig. 4. It can be readily seen that the relativistic effects strongly violate the nonrelativistic scaling rule stating that the function $\bar{R}(\omega) \equiv Z^2 R(\omega/\omega_{cr})$ does not depend on the nuclear charge. The relativistic enhancement of the ratio of the double-to-single photoionization cross sections can be conveniently parameterized as

$$R(\omega, Z) = R_{NR}(\omega, Z) \left[1 + (Z\alpha)^2 \frac{\omega}{\omega_{cr}} f_{rel}(\omega, Z) \right], \quad (26)$$

where R_{NR} is the nonrelativistic limit of the ratio σ^{++}/σ^+ and f_{rel} is a smooth function of the nuclear charge Z and photon energy ω . This function is plotted for several nuclear charges in Fig. 5. We observe that the numerical values of f_{rel} lay within the interval of (1.5, 3.5) for the wide range of nuclear charge numbers and photon energies. We also found that for $Z \lesssim 20$, $f_{rel} \approx 3$ and is nearly Z -independent. In particular, for helium and 1 keV photon energy ($\omega/\omega_{cr} \approx 13$), the above formula predicts the relativistic enhancement of about

0.8%, which is negligible at the current level of experimental precision.

The normalized energy distribution of the ejected electrons is shown in Fig. 6 for several nuclear charges and photon energies. Again, we observe that the scaled energy distribution does not depend on the nuclear charge in the nonrelativistic limit and that this scaling is violated by the relativistic effects. Note that in the case of the energy distribution, the differential cross section has to be scaled by the factor of $Z^2(Z\alpha)^2/\sigma^+$ and not by Z^2/σ^+ as for the total cross section; the additional factor is due to the fact that the integration interval in Eq. (15) is proportional to $(Z\alpha)^2$. We also confirm the known statement [4] that the highly asymmetric energy sharing between the two emitted electrons dominates at the high photon energies, with one of the electrons carrying away most of the photon energy.

VI. CONCLUSION

In this paper, we presented a relativistic calculation of the double photoionization of helium-like atoms. Our approach is based on the partial-wave representation of the Dirac continuum states and accounts for the retardation in the electron-electron interaction, the higher-order multipoles of the absorbed photon as well as the interaction of the electrons with the nucleus without any expansion in the binding field. The electron-electron interaction is taken into account to the first order of perturbation theory. The omitted higher-order electron correlation effects are estimated by comparing our numerical results for atomic helium with the experimental results and the available nonrelativistic theory. The calculational results are shown to be gauge invariant both with respect of the gauge of the absorbed photon and the gauge of the electron-electron interaction.

Our calculation shows that the relativistic effects become prominent in the double photoionization cross section already for medium- Z ions. These effects change the shape of the energy dependence of the ratio of the double-to-single photoionization $R(\omega)$ drastically. In particular, the well-known constant high-energy asymptotic behaviour of $R(\omega)$ in helium gives place to the monotonically

growing behaviour in the case of helium-like targets with $Z \gtrsim 20$.

We note that the reported magnitude of the relativistic effects is insufficient to explain the large discrepancy between theory and experiment observed for the double K -shell photoionization in neutral atoms. In particular, for the photon energy of 90 keV in silver ($\omega/\omega_{\text{cr}} = 1.73$), we obtain the relativistic enhancement of about 50% [see Eq. (26)], which is much smaller than the 4-fold disagreement between theory and experiment [13].

It should be mentioned that in the present investigation we do not consider the Compton scattering mechanism of the total double photoionization, which is known to contribute significantly for high-energy photons [5]. This reaction channel is more difficult for a relativistic calculation than the photoabsorption considered in the present work. To a certain extent, the photoabsorption and the Compton mechanisms of the double photoionization can be distinguished in the experiment by measuring the momentum vector of the recoil ions [41].

Acknowledgement

The work reported in this paper was supported by the Helmholtz Gemeinschaft (Nachwuchsgruppe VH-NG-421). The computations were partly performed on the computer cluster of St. Petersburg State Polytechnical University.

Appendix A: Single photoionization: calculation formulas

In order to perform integrations over the angular variables in the matrix element of the photon absorption operator, we fix the z -axis of our coordinate system to be directed along the photon momentum \mathbf{k} and assume that the polarization vector $\hat{\mathbf{u}}$ has the only nonvanishing spherical component u_λ ($\lambda = \pm 1$). The result for the matrix element of the gauge-independent part of the photon absorption operator is

$$\langle \kappa_a \mu_a | \boldsymbol{\alpha} \cdot \hat{\mathbf{u}}_\lambda e^{i\mathbf{k} \cdot \mathbf{r}} | \kappa_n \mu_n \rangle = (-1) \sum_{JL} i^{1+L} \Pi_L (-1)^{j_n - \mu_n} C_{j_a \mu_a, j_n - \mu_n}^{J\lambda} C_{L0, 1\lambda}^{J\lambda} P_{JL}(an), \quad (\text{A1})$$

where $\Pi_L = \sqrt{2L+1}$ and the initial and final states are the Dirac states with given relativistic angular momentum quantum number κ and the momentum projection μ . The radial integrals P_{JL} are

$$P_{JL}(an) = \int_0^\infty dr r^2 j_L(\omega r) [g_n(r) f_a(r) S_{JL}(\kappa_n, -\kappa_a) - f_n(r) g_a(r) S_{JL}(-\kappa_n, \kappa_a)], \quad (\text{A2})$$

where j_L is the spherical Bessel function, g_i and f_i are the upper and the lower radial components of the Dirac wave function, respectively, and S_{JL} are the angular coefficients given by Eqs. (C7)-(C9) of Ref. [42]. The matrix element

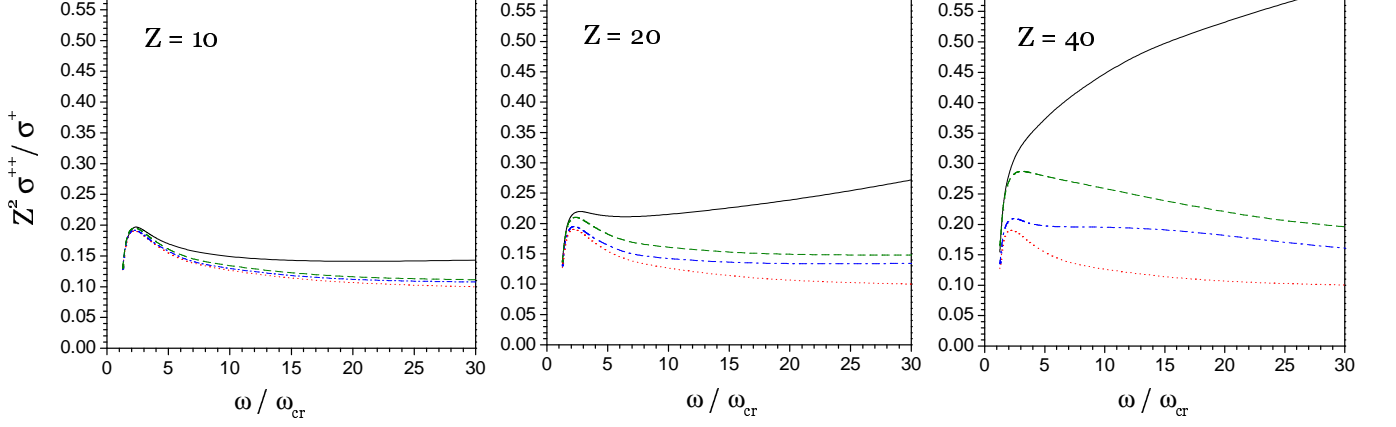


FIG. 3: (Color online) Comparison of the results of the fully relativistic calculations (solid line, black) with results of various approximate treatments. The dotted (red) line represents the nonrelativistic limit. The dashed-dotted (blue) line is obtained with the relativistic wave functions but without retardation and with including the relativistic $E1$ transition only. The dashed (green) line shows the results obtained with the relativistic wave functions and the full retardation but with including the relativistic $E1$ transition only. The threshold photon energy of the double photoionization is $\omega_{\text{cr}} = 2.56$ keV for Ne^{8+} , $\omega_{\text{cr}} = 10.60$ keV for Ca^{18+} , and $\omega_{\text{cr}} = 43.75$ keV for Zr^{38+} .

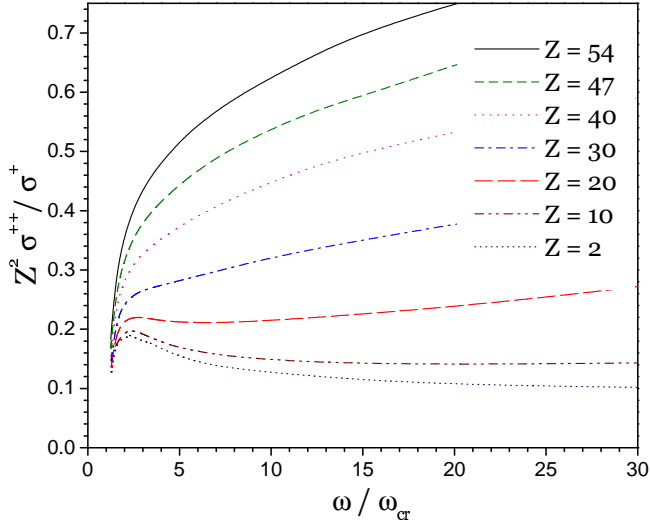


FIG. 4: (Color online) Relativistic results for the scaled ratio of the double photoionization cross section σ^{++} to the single photoionization cross section σ^+ , for helium-like ions with different nuclear charges Z , as a function of the photon energy ω divided by the threshold photon energy of the double photoionization ω_{cr} .

of the gauge-dependent part of the photon absorption operator can be evaluated as

$$\langle \kappa_a \mu_a | (\boldsymbol{\alpha} \cdot \hat{\mathbf{k}} - 1) e^{i\mathbf{k} \cdot \mathbf{r}} | \kappa_n \mu_n \rangle = (-1) \sum_{JL} i^L \Pi_L (-1)^{j_n - \mu_n} C_{j_a \mu_a, j_n - \mu_n}^{J0} \left[i C_{L0,10}^{J0} P_{JL}(an) + \delta_{J,L} C_L(\kappa_a, \kappa_n) R_L(an) \right], \quad (\text{A3})$$

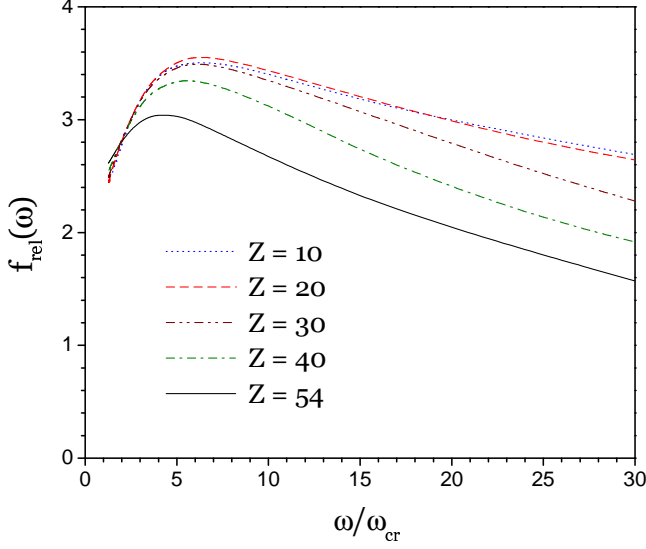


FIG. 5: (Color online) Relativistic enhancement function f_{rel} defined by Eq. (26), for different helium-like ions.

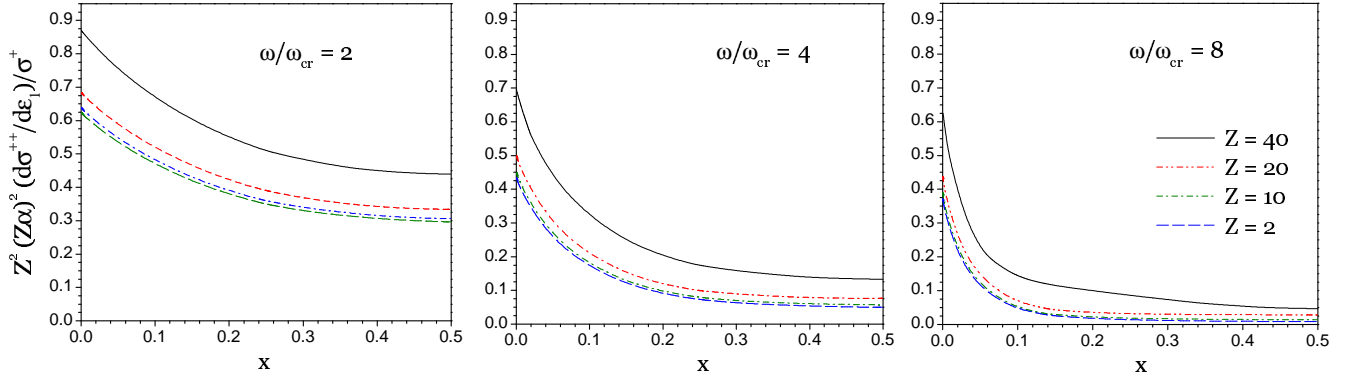


FIG. 6: (Color online) Normalized energy distribution of the ejected electrons as a function of the energy sharing parameter x defined by Eqs. (12) and (13), for different nuclear charge numbers and photon energies.

where the angular coefficients C_L are given by Eq. (C10) of Ref. [42] and R_L is the radial integral

$$R_L(an) = \int_0^\infty dr r^2 j_L(\omega r) [g_n(r) g_a(r) + f_n(r) f_a(r)]. \quad (\text{A4})$$

Appendix B: Double photoionization: calculation formulas

Let us write the amplitude (16) as

$$\tau_\lambda^{++} = \tau_{12ab} - \tau_{21ab} - \tau_{12ba} + \tau_{21ba}, \quad (\text{B1})$$

where τ_{12ab} corresponds to the part of Eq. (16) with $P\varepsilon_1 P\varepsilon_2 = \varepsilon_1 \varepsilon_2$ and $QaQb = ab$ and the remaining three terms are obtained by permutations. The contri-

butions due to the two terms in the braces of Eq. (16) will be denoted by subscripts A and B , respectively, $\tau_{12ab} = \tau_{12ab,A} + \tau_{12ab,B}$.

The calculation formulas take the simplest form in the case when the initial electron state is the ground state of the atom. In this case, the permutation over the a and b electrons yields just a combinatorial factor and all summations over the momentum projections can be evaluated in the closed form. The result for the single

differential cross section is

$$\frac{d\sigma^{++}}{d\varepsilon_1} = \frac{4\pi^2\alpha}{\omega} \sum_{\kappa_1\kappa_2J} \left| 2 \left[\tilde{\tau}_{12ab} + (-1)^{j_1-j_2+J} \tilde{\tau}_{21ab} \right] \right|^2. \quad (\text{B2})$$

The amplitude is $\tilde{\tau}_{12ab} = \tilde{\tau}_{12ab,A} + \tilde{\tau}_{12ab,B}$,

$$\tilde{\tau}_{12ab,A} = \frac{\alpha}{\sqrt{2}} \sum_{LL'\kappa_n} (-1)^{1+L} \Pi_L C_{L0,1\lambda}^{J\lambda} \frac{(-1)^{L'+J+j_n+j_2}}{\sqrt{2}} \left\{ \begin{matrix} j_1 & j_2 & J \\ \frac{1}{2} & j_n & L' \end{matrix} \right\} \sum_n \frac{R_{L'}(\Delta, \varepsilon_1 \varepsilon_2 n b) P_{JL}(na)}{\varepsilon_a + \omega - \varepsilon_n}, \quad (\text{B3})$$

and

$$\tilde{\tau}_{12ab,B} = \frac{\alpha}{\sqrt{2}} \sum_{LL'\kappa_n} (-1)^{1+L} \Pi_L C_{L0,1\lambda}^{J\lambda} \frac{\delta_{j_n,j_2}}{\sqrt{2}(2j_2+1)} \sum_n \frac{P_{JL}(\varepsilon_1 n) R_{L'}(\Delta, n \varepsilon_2 ab)}{\varepsilon_1 - \omega - \varepsilon_n}, \quad (\text{B4})$$

where $R_L(\Delta, abcd)$ is the relativistic generalization of the Slater integral for the electron-electron interaction given

by Eqs. (C1)-(C10) of Ref. [42] and $\Delta = \varepsilon_2 - \varepsilon_b$.

-
- [1] J. S. Briggs and V. Schmidt, J. Phys. B **33**, R1 (2000).
 - [2] H. Kossmann, V. Schmidt, and T. Andersen, Phys. Rev. Lett. **60**, 1266 (1988).
 - [3] F. Maubetsch and J. S. Briggs, Phys. Rev. Lett. **68**, 2004 (1992).
 - [4] M. Y. Amucia, E. G. Drukarev, V. G. Gorshkov, and M. P. Kazachkov, J. Phys. B **8**, 1248 (1975).
 - [5] L. R. Andersson and J. Burgdörfer, Phys. Rev. Lett. **71**, 50 (1993).
 - [6] R. C. Forrey, H. R. Sadeghpour, J. D. Baker, J. D. Morgan, and A. Dalgarno, Phys. Rev. A **51**, 2112 (1995).
 - [7] J.-Z. Tang and I. Shimamura, Phys. Rev. A **52**, R3413 (1995).
 - [8] A. S. Kheifets and I. Bray, Phys. Rev. A **54**, R995 (1996).
 - [9] K. W. Meyer, C. H. Greene, and B. D. Esry, Phys. Rev. Lett. **78**, 4902 (1997).
 - [10] Y. Qiu, J.-Z. Tang, J. Burgdörfer, and J. Wang, Phys. Rev. A **57**, R1489 (1998).
 - [11] E. P. Kanter, R. W. Dunford, B. Krässig, and S. H. Southworth, Phys. Rev. Lett. **83**, 508 (1999).
 - [12] S. H. Southworth, E. P. Kanter, B. Krässig, L. Young, G. B. Armen, J. C. Levin, D. L. Ederer, and M. H. Chen, Phys. Rev. A **67**, 062712 (2003).
 - [13] E. P. Kanter, I. Ahmad, R. W. Dunford, D. S. Gemmell, B. Krässig, S. H. Southworth, and L. Young, Phys. Rev. A **73**, 022708 (2006).
 - [14] J. Hoszowska, A. K. Kheifets, J.-C. Dousse, M. Berset, I. Bray, W. Cao, K. Fennane, Y. Kayser, M. Kavčič, J. Szlachetko, and M. Szlachetko, Phys. Rev. Lett. **102**, 073006 (2009).
 - [15] J. Hoszowska, J.-C. Dousse, W. Cao, K. Fennane, Y. Kayser, M. Szlachetko, J. Szlachetko, and M. Kavčič, Phys. Rev. A **82**, 063408 (2010).
 - [16] S. W. Epp, J. R. C. López-Urrutia, G. Brenner, V. Mäkel, P. H. Mokler, R. Treusch, M. Kuhlmann, M. V. Yurkov, J. Feldhaus, J. R. Schneider, M. Wellhöfer, M. Martins, W. Wurth, and J. Ullrich, Phys. Rev. Lett. **98**, 183001 (2007).
 - [17] A. Simon, A. Warczak, T. ElKafrawy, and J. A. Tanis, Phys. Rev. Lett. **104**, 123001 (2010).
 - [18] G. Bednarz, D. Sierpowski, T. Stöhlker, A. Warczak, H. Beyer, F. Bosch, A. Bräuning-Demian, H. Bräuning, X. Cai, A. Gumberidze, S. Hagmann, C. Kozhuharov, D. Liesen, X. Ma, P. Mokler, A. Muthig, Z. Stachura, and S. Toleikis, Nucl. Instrum. Methods **B 205**, 573 (2003), 11th International Conference on the Physics of Highly Charged Ions.
 - [19] J. Eichler and W. Meyerhof, *Relativistic Atomic Collisions*, Academic Press, San Diego, 1995.
 - [20] M. E. Rose, *Relativistic Electron Theory*, John Wiley & Sons, NY, 1961.
 - [21] J. Eichler and T. Stöhlker, Phys. Rev. **439**, 1 (2007).
 - [22] A. Mikhailov, A. Nefiodov, and G. Plunien, Phys. Lett. A **358**, 211 (2006).
 - [23] V. M. Shabaev, Phys. Rep. **356**, 119 (2002).
 - [24] A. I. Mikhailov, I. A. Mikhailov, A. N. Moskaev, A. V. Nefiodov, G. Plunien, and G. Soff, Phys. Rev. A **69**, 032703 (2004).
 - [25] V. M. Shabaev, V. A. Yerokhin, T. Beier, and J. Eichler, Phys. Rev. A **61**, 052112 (2000).
 - [26] V. A. Yerokhin, V. M. Shabaev, T. Beier, and J. Eichler, Phys. Rev. A **62**, 042712 (2000).
 - [27] V. A. Yerokhin and A. Surzhykov, Phys. Rev. A **81**, 062703 (2010).
 - [28] P. J. Mohr, G. Plunien, and G. Soff, Phys. Rep. **293**, 227 (1998).
 - [29] V. A. Yerokhin and A. Surzhykov, Phys. Rev. A **82**, 062702 (2010).
 - [30] E. Caliceti, M. Meyer-Hermann, P. Ribeca, A. Surzhykov, and U. Jentschura, Phys. Rep. **446**, 1 (2007).
 - [31] M. A. Kornberg and J. E. Miraglia, Phys. Rev. A **49**,

- 5120 (1994).
- [32] A. I. Mikhailov, A. V. Nefiodov, and G. Plunien, *J. Phys. B* **42**, 231003 (2009).
 - [33] A. I. Mikhailov and I. A. Mikhailov, *Zh. Eksp. Teor. Fiz.* **114**, 1537 (1998), [*Sov. Phys. JETP* **87**, 833 (1998)].
 - [34] J. C. Levin, G. B. Armen, and I. A. Sellin, *Phys. Rev. Lett.* **76**, 1220 (1996).
 - [35] R. Dörner, T. Vogt, V. Mergel, H. Khemliche, S. Kravis, C. L. Cocke, J. Ullrich, M. Unverzagt, L. Spielberger, M. Damrau, O. Jagutzki, I. Ali, B. Weaver, K. Ullmann, C. C. Hsu, M. Jung, E. P. Kanter, B. Sonntag, M. H. Prior, E. Rotenberg, J. Denlinger, T. Warwick, S. T. Manson, and H. Schmidt-Böcking, *Phys. Rev. Lett.* **76**, 2654 (1996).
 - [36] J. C. Levin, I. A. Sellin, B. M. Johnson, D. W. Lindle, R. D. Miller, N. Berrah, Y. Azuma, H. G. Berry, and D.-H. Lee, *Phys. Rev. A* **47**, R16 (1993).
 - [37] J. A. R. Samson, W. C. Stolte, Z.-X. He, J. N. Cutler, Y. Lu, and R. J. Bartlett, *Phys. Rev. A* **57**, 1906 (1998).
 - [38] C. J. Bostock, D. V. Fursa, and I. Bray, *Phys. Rev. A* **80**, 052708 (2009).
 - [39] A. I. Mikhailov, I. A. Mikhailov, A. N. Moskalev, A. V. Nefiodov, G. Plunien, and G. Soff, *Phys. Lett. A* **316**, 395 (2003).
 - [40] V. M. Shabaev, I. I. Tupitsyn, V. A. Yerokhin, G. Plunien, and G. Soff, *Phys. Rev. Lett.* **93**, 130405 (2004).
 - [41] L. Spielberger, O. Jagutzki, R. Dörner, J. Ullrich, U. Meyer, V. Mergel, M. Unverzagt, M. Damrau, T. Vogt, I. Ali, K. Khayyat, D. Bahr, H. G. Schmidt, R. Frahm, and H. Schmidt-Böcking, *Phys. Rev. Lett.* **74**, 4615 (1995).
 - [42] V. A. Yerokhin and V. M. Shabaev, *Phys. Rev. A* **60**, 800 (1999).